

A Study on the Irradiation Effects of Radioactivity for the Molding of Polymer

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This paper deals with an empirical study of the effect of irradiation of Co^{60} γ rays on molding of polyethylene with respect to cross-linking, where different atmospheric conditions of irradiation of γ rays are employed.

Introduction

Irradiation of radioactivity to polymers causes chemical reactions which contribute to the study of molding of polymers.

While radioactivity passes through a polymer, molecules and atoms become excited and most of the incident energy is provided for the polymer with the corresponding loss of its own energy, the electrons being sprung out. This chain reaction excites molecules, ions and electrons which cause complicate reactions among the neighbouring molecules. Then ions in the molecules are neutralized, excited molecules decomposed, free radicals produced. These free radicals react among themselves or with molecules. It can be assumed that in this reaction the microstructure of the polymer undergoes some changes and part of the polymer takes a reticulate structure of three dimensions.

The five different quantities of γ rays ($1 \times 10^6 \text{r}$, $5 \times 10^6 \text{r}$, $1 \times 10^7 \text{r}$, $5 \times 10^7 \text{r}$ and $1 \times 10^8 \text{r}$, respectively) were applied in both air and water in the atmosphere of 20°C , 50°C , and 80°C , to the injection-forming pellets of the high-pressured polyethylene.

Then the pellets at hot pressures were heated at the heating velocity of 110°C/hr from 60°C to 170°C and compressed at the degree of 200kg/cm^2 . After that they were kept at 170°C for 10 minutes and cooled down to 60°C in about 2 hours for molding. The molding characteristic was estimated by the use of stress-strain diagrams obtained by tension tests with autograph, where specimens of Dumb-bell shape (JIS K 6475 Type) were used.

Experimental Results and Consideration

The experimental results are illustrated in Fig. 1 to Fig. 6, where each number of the curves represents the quantity of irradiation of radioactivity : 1 is for $1 \times 10^6 \text{r}$, 2 for $5 \times 10^6 \text{r}$, 3 for $1 \times 10^7 \text{r}$, 4 for $5 \times 10^7 \text{r}$, 5 for $1 \times 10^8 \text{r}$.

Fig. 1 to Fig. 3 show the molding in case of the irradiation in air. Here care must be taken to the common feature in the effects of irradiation of radioactivity upon molding:

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The cases of 1×10^6 r, 5×10^6 r, 1×10^7 r can be grouped into one category as they show similar characteristics, while those of 5×10^7 r and 1×10^8 r can be grouped into another. This implies that the microstructure of polyethylene is quite different from that of 5×10^7 r suggesting different productions of free radicals as well as different reactions of free radicals among themselves or with molecules. Difference can be also found in the structure of cross linking. Therefore the polyethylene irradiated above 5×10^7 r of radioactivity decreases in fluidity and tends to be lack in both molding and autohesion among molecules. Moreover in each atmosphere of irradiation temperature elongation

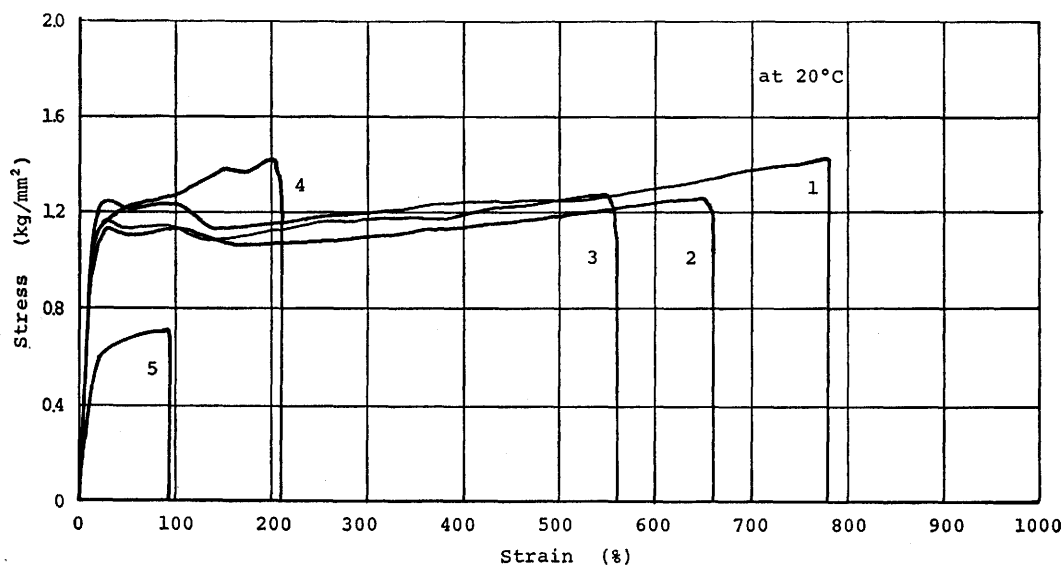


Fig. 1 Stress-strain diagrams of hot pressed specimens after the irradiation in air

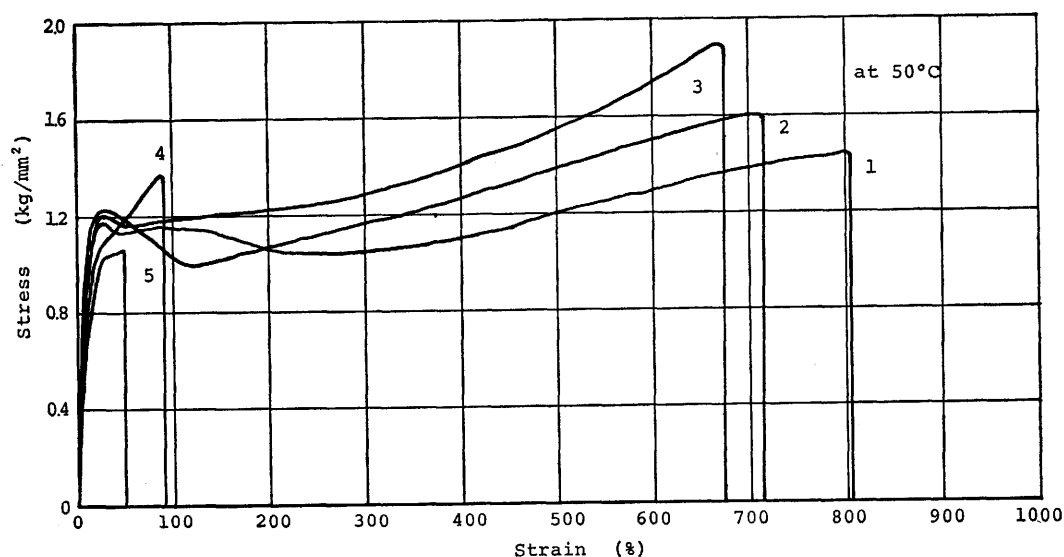


Fig. 2 Stress-strain diagrams of hot pressed specimens after the irradiation in air

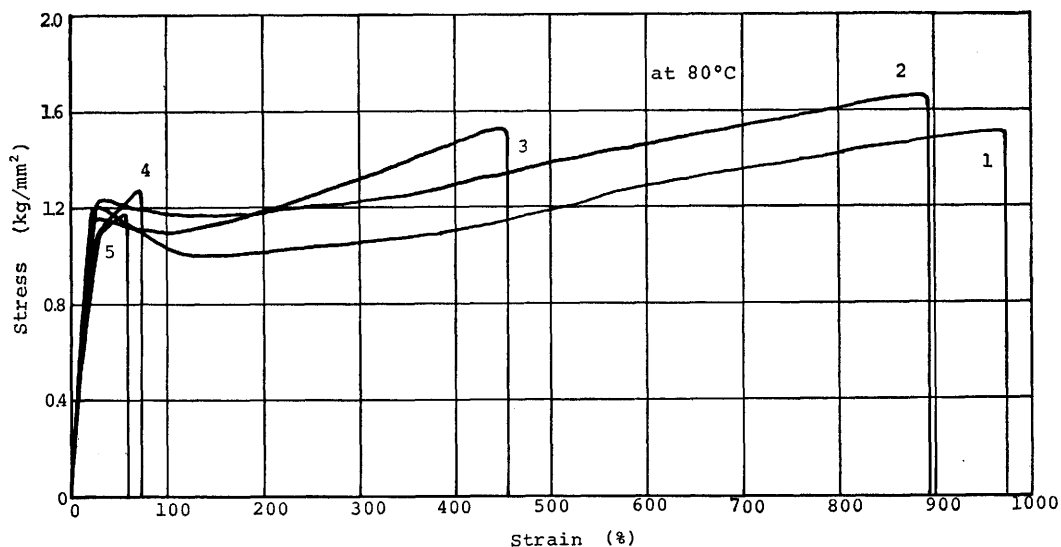


Fig. 3 Stress-strain diagrams of hot pressed specimens after the irradiation in air

of each specimen up to the breaking point decreases with increase of the quantity of radioactivity, while from the beginning to the breaking point the absorbed energy decreases with increase of radioactivity, not so much difference being found in the breaking strength. This explains the fact that irradiation of radioactivity changes the structure of materials to a brittle from which was most strikingly seen in the case of above 5×10^7 r of γ rays. When irradiation is done at above the transition temperature (80°C), molecules can move rather easily and at the same time the production of free radicals as well as the reaction among themselves can be easily done.

As in Fig. 3, there can be found many changes of molding in the case of 1×10^7 r. As for 5×10^7 r and 1×10^8 r brittleness is more conspicuously seen in the irradiation at 80°C . The above results suggest the following.

- 1) Elongation of materials decreases with quantity of radioactivity. Especially its remarkable change can be seen between 1×10^7 r and 5×10^7 r.
- 2) The absorbed energy changes strikingly till the materials reach their breaking points.
- 3) The materials which were molded after the irradiation of 1×10^6 r to 1×10^8 r did not change so greatly their breaking strengths.
- 4) The irradiation at above the transition temperature effects the molded materials more conspicuously suggesting their great influence upon the fluidity and molding of polymers. Let us take one experiment performed by Lawton et al. who examined the effects of the irradiation temperature from -170°C to $+240^\circ\text{C}$ on such polyethylenes as will cross-link even at normal temperatures. This experiment shows that the higher the temperature is, the more easily occurs cross-linking.

Fig. 7¹⁾ illustrates the effects of temperature on the production of gels when 1.23×10^7 r of radioactivity was equally irradiated Marlex 50's. Rapidly increased effects on the

cross-linking can be found between the glass transition temperature ($T_g = -40^\circ\text{C}$) and melting point ($T_m = 135^\circ\text{C}$). This well corresponds to the author's experimental results with respect to fluidity and molding which were performed in the atmospheres of 20°C , 50°C and 80°C .

Fig. 4 to Fig. 6 illustrate the stress-strain diagrams obtained by the tension tests with autograph IM 100. The specimens were made from films produced by first irradiation of Co^{60} γ rays at 20°C , 50°C and 80°C in water, and then by hot-pressing in the same condition. The numbers in the diagrams represent the quantity of irradiation as in Fig. 1 to Fig. 3. In these experimental results comparison with the cases of irradiation

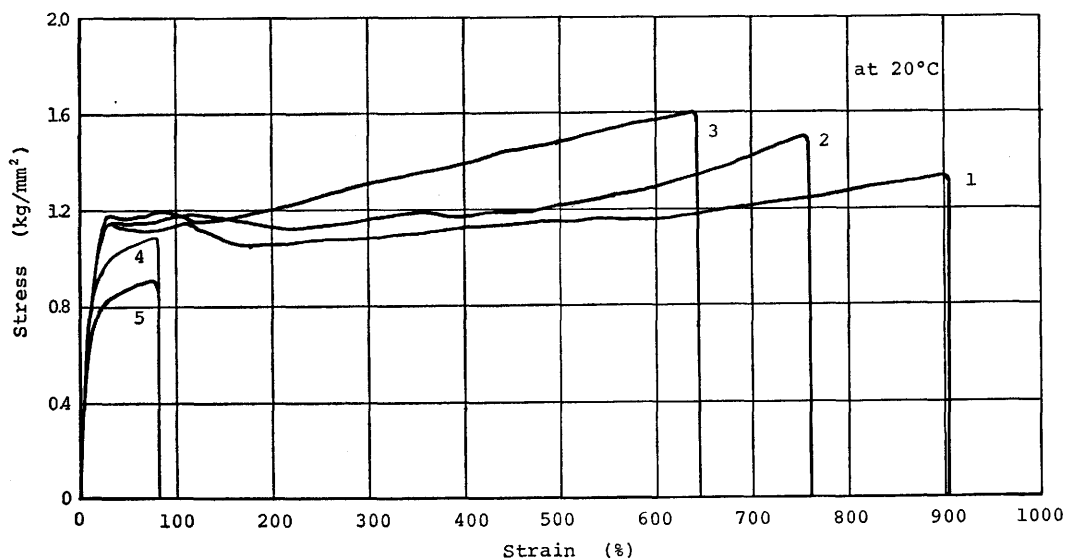


Fig. 4 Stress-strain diagrams of hot pressed specimens after the irradiation in water

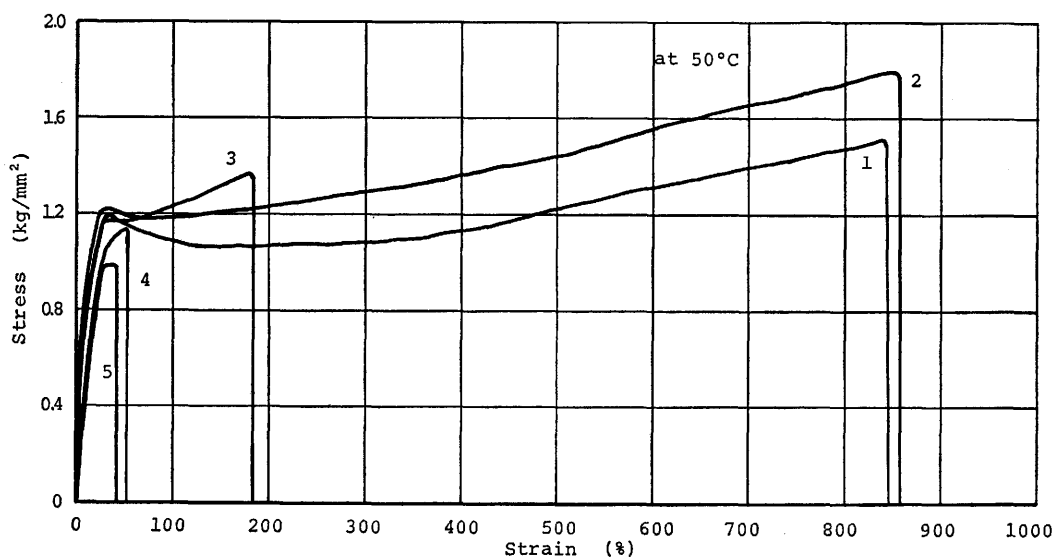


Fig. 5 Stress-strain diagrams of hot pressed specimens after the irradiation in water

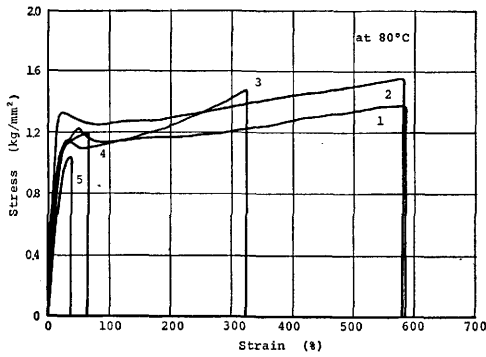


Fig. 6 Stress-strain diagrams of hot pressed specimens after the irradiation in water

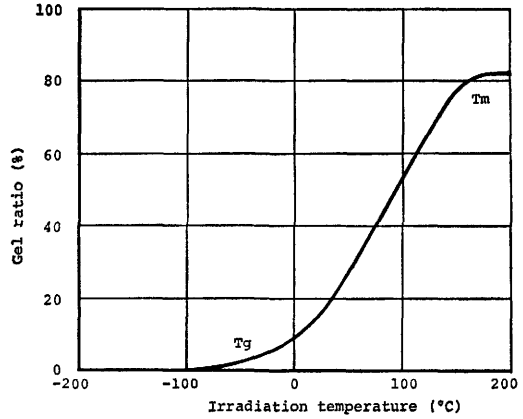


Fig. 7 Relation between gel ratio and irradiation temperature of radioactivity to polyethylene Marlex 50 (by Lawton)

in air presents the clear difference that as in Fig. 6 both elongation and absorbed energy up to the breaking point show rapid decreases tending to become brittle. This may be explained as follows: Above the transition temperature segments of polyethylene can easily move. Moreover absorption of 0.01 percentage of the water per 24 hours causes decomposition of water in irradiation both on and within the polyethylene, producing H radicals and OH radicals which facilitate the motion of molecules in each part of them. It may be assumed that due to these reactions cross-linking occurs

Table 1 Tensile strength of Irradiated specimens at 20°C

Co ⁶⁰ γ-ray	Atmosphere : In air			Atmosphere : In water		
	No. 1	No. 2	No. 3	No. 1	No. 2	No. 3
1×10^6	1.17	1.24	1.45	1.19	1.31	1.15
5×10^6	1.13	1.18	1.22	1.16	1.15	1.16
1×10^7	1.16	1.16	1.16	1.18	1.17	1.07
5×10^7	1.22	1.26	1.21	0.97	1.01	1.03
1×10^8	0.57	0.70	0.72	0.78	0.80	0.84

Table 2 Tensile strength of Irradiated specimens at 50°C

Co ⁶⁰ γ-ray	Atmosphere : In air			Atmosphere : In water		
	No. 1	No. 2	No. 3	No. 1	No. 2	No. 3
1×10^6	1.18	1.28	1.19	1.23	1.19	1.26
5×10^6	1.43	1.22	1.22	1.22	1.22	1.22
1×10^7	1.18	1.43	1.19	1.17	1.18	1.17
5×10^7	1.08	1.15	1.29	1.05	1.05	1.04
1×10^8	1.13	1.02	1.07	1.15	1.01	0.98

Table 3 Tensile strength of Irradiated specimens at 80°C

Co ⁶⁰ γ-ray	Atmosphere : In air			Atmosphere : In water		
	No. 1	No. 2	No. 3	No. 1	No. 2	No. 3
1 × 10 ⁶	1.20	1.20	1.22	1.23	1.20	1.22
5 × 10 ⁶	1.23	1.22	1.23	1.32	1.25	1.17
1 × 10 ⁷	1.15	1.33	1.15	1.17	1.14	1.17
5 × 10 ⁷	1.15	1.17	1.23	1.13	1.07	1.10
1 × 10 ⁸	1.11	1.11	1.12	0.98	0.90	1.03

more easily. Therefore is it not pertinent to induce that irradiation of radioactivity in water in the atmosphere of high temperature has great effects on the fluidity and molding of polyethylene? The above experiments were performed with three specimens in the same condition. Fig. 1 to Fig. 6 show each case of the various conditions. Of all the experimental results yielding strengths are listed in Table 1 to Table 3 which suggest that the quantitative effect of irradiation is relatively small. On performing the same experiments as stated above, F. R. T. P's were produced by hot-pressing.

Then using autographs fluidity and molding were studied.

The results are listed in Table 4 to Table 6. Here the tensile strength representing

the yielding strength is by far greater than that of straight polyethylene.²⁾ This experiment aims at examining the possibility of F. R. T. P, by only quantitatively studying the adhesive property for glass fibers and their structures and contents. In this experiment cloths of glass fiber for F. R. T. P were used and their contents were 30% (wt.).

Table 4 Tensile strength of Irradiated F. R. T. P. specimens at 20°C

Atmosphere Co ⁶⁰ γ-ray	In air	In water
1 × 10 ⁶	13.06	11.50
5 × 10 ⁶	12.16	8.81
1 × 10 ⁷	10.37	5.95
5 × 10 ⁷	9.38	6.23
1 × 10 ⁸	7.94	5.93

Table 5 Tensile strength of Irradiated F. R. T. P. specimens at 50°C

Atmosphere Co ⁶⁰ γ-ray	In air	In water
1 × 10 ⁶	13.27	14.04
5 × 10 ⁶	10.85	13.19
1 × 10 ⁷	8.76	7.82
5 × 10 ⁷	8.49	5.18
1 × 10 ⁸	13.29	4.17

Table 6 Tensile strength of Irradiated F. R. T. P. specimens at 80°C

Atmosphere Co ⁶⁰ γ-ray	In air	In water
1 × 10 ⁶	17.96	14.66
5 × 10 ⁶	14.21	10.70
1 × 10 ⁷	11.08	10.48
5 × 10 ⁷	8.72	7.86
1 × 10 ⁸	10.00	6.71

Conclusion

The following summary can be made from the results of the present experiment.

- 1) Elongation of molded materials decrease and becomes brittle with increase of the quantity of radioactivity.
- 2) A rapid change of fluidity and molding can be found between $1 \times 10^7 \text{r}$ and $5 \times 10^7 \text{r}$.
- 3) The quantity of radioactivity has a relatively slight effect on strength.
- 4) With increase of the atmosphere of irradiation temperature, fluidity and molding decrease.
- 5) The effect of irradiation in the atmosphere of high temperatures is, if in water, much greater.
- 6) It is possible to mold F. R. T. P's from the irradiated polyethylene. They have great strength.

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